

①9 RÉPUBLIQUE FRANÇAISE
INSTITUT NATIONAL
DE LA PROPRIÉTÉ INDUSTRIELLE
PARIS

①1 N° de publication : **2 610 635**
(à n'utiliser que pour les
commandes de reproduction)
②1 N° d'enregistrement national : **87 01623**

⑤1 Int Cl⁴ : C 08 L 75/06; B 29 C 43/24, 45/10, 47/14;
A 61 G 17/00; B 29 D 7/01 // (C 08 L 75/06, 1:00, 3:02).

⑫

DEMANDE DE BREVET D'INVENTION

A1

②2 Date de dépôt : 10 février 1987.

③0 Priorité :

④3 Date de la mise à disposition du public de la
demande : BOPI « Brevets » n° 32 du 12 août 1988.

⑥0 Références à d'autres documents nationaux appa-
rentés :

⑦1 Demandeur(s) : *Société anonyme dite : SOCIÉTÉ FRAN-
CAISE D'EXTRUSION ET DE CALANDRAGE (S.F.E.C.) et
Etablissement Public dit : CHARBONNAGES DE FRANCE.*
— FR.

⑦2 Inventeur(s) : Jean-Marc Aime; Georges Mention; An-
dré Thouzeau.

⑦3 Titulaire(s) :

⑦4 Mandataire(s) : Rinuy et Santarelli.

⑤4 Compositions à base de polyuréthane permettant l'obtention par calandrage de matériaux biodégradables.

⑤7 Ces compositions comprennent en plus de la résine po-
lyuréthane qui en est le constituant essentiel, un hydrate de
carbone, au moins un deuxième polymère thermoplastique
miscible à l'état fondu avec le polyuréthane et/ou au moins
un troisième polymère biodégradable choisi parmi les polyesters
aliphatiques biodégradables, de préférence de même nature
que la base polyester du polyuréthane mis en œuvre.

FR 2 610 635 - A1

La présente invention concerne des feuilles,
5 films ou plaques biodégradables à base de polyuréthane,
ainsi que leur obtention par calandrage.

On a déjà proposé de fabriquer des feuilles
biodégradables par extrusion soufflage de mélanges de
polyuréthane et d'amidon (brevet britannique
10 N° 1.524.821). Toutefois, de tels mélanges ne peuvent
être soumis à la technique du calandrage dans des con-
ditions de productivité acceptables. En outre, la biodé-
gradabilité et les propriétés mécaniques des feuilles
obtenues peuvent être insuffisantes, en fonction de la
15 nature du polyuréthane utilisé.

La présente invention a pour but de proposer
des feuilles, films ou plaques (dits ci-après "pro-
duits") à base de polyuréthane qui aient de bonnes
qualités mécaniques et de biodégradabilité et qui
20 puissent être obtenus par calandrage, dans les
conditions qui sont habituellement mises en oeuvre par
exemple pour la fabrication des feuilles de PVC souples
(débit machine comparable, ligne classique de calan-
drage, etc.).

25 L'invention concerne par conséquent notamment
les compositions à mettre en oeuvre pour l'obtention de
ces matériaux.

Selon l'invention, ces compositions compren-
nent, en plus de la résine polyuréthane qui en est le
30 constituant essentiel, un hydrate de carbone, au moins
un deuxième polymère thermoplastique miscible à l'état
fondu avec le polyuréthane et/ou au moins un troisième
polymère biodégradable choisi parmi les polyesters ali-
phatiques biodégradables, de préférence de même nature
35 que la base polyester du polyuréthane mis en oeuvre.

De préférence, le deuxième polymère utilisé est du PVC ou un mélange de PVC et d'un plastifiant monomérique ou polymérique du PVC.

Le plastifiant est par exemple un phtalate, tel que le dioctylphtalate.

Ce deuxième polymère éventuellement plastifié, lorsqu'il n'est pas lui-même biodégradable, est de préférence mis en oeuvre en quantité aussi faible que possible, si la biodégradabilité est la principale qualité recherchée. Cette proportion sera fonction du taux d'hydrate de carbone introduit, par rapport auquel elle pourra représenter par exemple 25 à 100 % en poids.

Le polyuréthane mis en oeuvre est choisi parmi les polyuréthanes thermoplastiques aliphatiques à base polyester. La base polyester sera choisie de préférence parmi les bases des composés ayant la meilleure biodégradabilité, par exemple la poly- ϵ -caprolactone, la poly- β -caprolactone. Par exemple, on utilisera un polyuréthane à base polycaprolactone.

De façon avantageuse, une composition selon l'invention associera une polycaprolactone (troisième polymère) à un polyuréthane à base polycaprolactone.

Ainsi, le troisième polymère, lorsqu'il est présent dans la composition de l'invention, le sera dans une proportion de 0 à 20 % en poids par rapport au mélange global.

On pourra également utiliser le poly- β -hydroxybutyrate et le polyhydroxyvalérate.

L'hydrate de carbone est un composé tel que fécule, amidon, amidon modifié, cellulose, dérivés de la cellulose, dextrine, lactose, manitol, lignine et il est utilisé en proportion de 10 à 45% en poids par rapport au mélange global.

Les Demandeurs ont constaté que la présence du troisième polymère peut faciliter la mise en oeuvre, augmenter la biodégradabilité et les propriétés mécaniques des produits obtenus.

Les compositions selon l'invention comprennent en outre les additifs classiques utilisés dans l'industrie du calandrage : lubrifiants, pigments, colorants, stabilisants, antioxydants, antibloquants, etc., bien connus de l'homme de l'art.

Les compositions selon l'invention sont mises en oeuvre par la technique classique du calandrage, telle qu'elle est utilisée pour la fabrication des feuilles de PVC. Il est à noter que l'appareillage utilisé pour le PVC peut être mis en oeuvre sans modification et avec des débits de production équivalents.

D'autres techniques de fabrication connues peuvent être également utilisées éventuellement à la place du calandrage pour la fabrication de feuilles, films ou plaques équivalentes, par exemple l'extrusion calandrage par filière plate, l'injection, etc.

Les produits selon l'invention ont des propriétés qui sont naturellement fonction de la composition de la formulation.

La biodégradabilité totale en poids peut atteindre 96 %. La vitesse de biodégradation déterminée selon la norme AFNOR NFX 41-514 (enfouissement dans un sol normalisé à 30°C, 95 % d'humidité relative, sur une feuille de 0,15 mm d'épaisseur) conduit à une perte totale des caractéristiques mécaniques de la feuille en un temps qui peut atteindre moins d'un mois.

Les produits obtenus sont élastiques, soudables, y compris par haute fréquence. Ils présentent un toucher agréable et doux, dû à l'association polyuréthane-hydrate de carbone.

Ils ont des caractéristiques mécaniques convenables pour leur utilisation, dans le domaine de l'emballage par exemple.

Ils sont stables au stockage dans les conditions normales.

Ces produits selon l'invention peuvent être utilisés :

- comme enveloppes mortuaires en conformité avec la législation française (housse, linceul et capitonage des cercueils) ;

- comme films agricoles ;
- 5 - comme films d'emballage souples ;
- comme matériaux de remplissage ou de rembourrage.

Exemple 1

On mélange à température ambiante, dans un mélangeur à pales :

- d'une part 40 parties de polyuréthane à base polycaprolactone et 13,5 parties de polycaprolactone ;
- d'autre part, 13,5 parties de PVC, 5,5 parties de dioctylphthalate, 27 parties de fécule de pomme de terre et 0,5 partie d'un stabilisant organométallique.

L'homogénéisation des deux mélanges est ensuite réalisée dans un mélangeur interne à 160°C pendant quelques minutes.

La mise en oeuvre est effectuée par calandrage sur une calandre à 4 cylindres, les températures des cylindres étant comprises entre 160 et 120°C.

On fabrique ainsi un film de 0,12 mm d'épaisseur et 2 mètres de largeur avec une vitesse linéaire de 30 mètres par minute.

1) Les caractéristiques mécaniques du film sont les suivantes :

a) caractéristiques mécaniques, en traction à 23°C, suivant norme NFT 54-102 :

- . module à 100 % d'allongement : 9 MPa,
- . force à la rupture : 15 MPa,
- . allongement à la rupture : 250 % ;

b) résistance à la déchirure à 23°C, suivant normes NFT 46-007 et ISO 34 :

- . éprouvette en croissant, déchirure amorcée : à une vitesse d'étirement de 50 mm/mn :
 - dans le sens trame : 6 MPa,
 - dans le sens chaîne : 7 MPa ;

à une vitesse d'étirement de 500 mm/mn :

- dans le sens trame : 7,5 MPa,

- dans le sens chaîne : 9 MPa ;

épreuve en croissant, déchirure non amor-

5 cée :

à une vitesse d'étirement de 50 mm/mn :

- dans le sens trame : 22 MPa,

- dans le sens chaîne : 25 MPa ;

à une vitesse d'étirement de 500 mm/mn :

10 - dans le sens trame : 12 MPa,

- dans le sens chaîne : 12 MPa ;

épreuve angulaire, déchirure amorcée :

- dans le sens trame : 6 MPa,

- dans le sens chaîne : 6 MPa.

15 2) Biodégradabilité

Suivant la norme NFX 41-514, on observe une diminution des propriétés mécaniques du film, plus exactement :

20 - au bout d'une semaine, le film est envahi totalement par des moisissures (changement de coloration). Par un examen plus approfondi au microscope à balayage électronique, on remarque la formation très nette de réseaux tridimensionnels qui correspondent à l'envahissement par les moisissures ;

25 - au bout de 15 jours d'enfouissement, un examen en microscope à balayage électronique montre la formation de microfissurations, caractéristiques de la dégradation du polyuréthane ;

30 - au bout de 10 semaines, l'échantillon laisse apparaître des fissurations visibles à l'oeil nu. L'échantillon est alors fragmenté et a perdu toutes ses propriétés mécaniques.

Exemple 2

35 Dans le but de réaliser une feuille présentant une biodégradabilité accrue, on opère comme dans l'exemple 1, sauf que l'on utilise les proportions suivantes :

- polyuréthane : 40 parties,
- polycaprolactone : 18 parties,
- PVC : 4,5 parties,
- DOP : 4 parties,
- 5 - féculé : 33 parties,
- stabilisant : 0,5 partie.

Sur la feuille obtenue, on effectue un essai de biodégradabilité suivant la norme NFX 41-514 :

- au bout de 3 jours, on observe un changement
- 10 de coloration caractérisant le début d'envahissement par les moisissures ;
- au bout d'une semaine, la feuille est totalement envahie par les moisissures et présente une coloration très sombre ;
- 15 - au bout de deux semaines, l'échantillon laisse apparaître des fragmentations mécaniques visibles à l'oeil nu avec un accroissement de la microfaune environnante ;
- au bout de quatre semaines, on observe une
- 20 disparition partielle de la matière de l'échantillon et, par conséquent, la perte totale des propriétés mécaniques.

Exemple 3

- On opère comme dans l'exemple 1, en utilisant
- 25 les proportions suivantes :
 - polyuréthane : 40 parties,
 - polycaprolactone : 25 parties,
 - PVC : 5 parties,
 - DOP : 4 parties,
 - 30 - féculé : 25 parties,
 - autres additifs : 1 partie.

Sur la feuille obtenue, on détermine la biodégradabilité, d'une part dans les conditions ordinaires, d'autre part en étirant le film dans sa limite de

35 réponse élastique. On observe une fragmentation à l'échelle microscopique analysée au microscope à balayage électronique, qui passe de 10 semaines à 5 semaines lorsque le film est sous tension.

Cet essai montre que, même avec une composition avec une forte teneur en matériau polymérique, la vitesse de dégradation reste élevée.

- 5 Il va de soi que la présente invention n'a été décrite qu'à titre illustratif et nullement limitatif et que toute modification, notamment au niveau des équivalences techniques, pourra y être apportée sans sortir de son cadre.

7. Compositions selon l'une quelconque des revendications 1 à 6, caractérisées en ce qu'elles associent une polycaprolactone (troisième polymère) à un polyuréthane à base polycaprolactone.

5 8. Compositions selon l'une quelconque des revendications 1 à 7, caractérisées en ce que ledit troisième polymère, lorsqu'il est présent, l'est dans une proportion de 0 à 20 % en poids par rapport au mélange global.

10 9. Compositions selon l'une quelconque des revendications 1 à 6, caractérisées en ce qu'elles associent au polyuréthane un poly- β -hydroxybutyrate ou un polyhydroxyvalérate.

15 10. Application des compositions selon l'une quelconque des revendications 1 à 9 à l'obtention de feuilles, films ou plaques biodégradables.

11. Application des compositions selon l'une quelconque des revendications 1 à 9 à l'obtention de feuilles, films ou plaques biodégradables par calandrage.

20 12. Feuilles, films ou plaques biodégradables obtenus à partir des compositions selon l'une quelconque des revendications 1 à 9.

25 13. Application des feuilles, films, plaques biodégradables selon la revendication 12 comme enveloppes mortuaires, films agricoles, films d'emballage souples, matériaux de remplissage ou de rembourrage.

7. Compositions selon l'une quelconque des revendications 1 à 6, caractérisées en ce qu'elles associent une polycaprolactone (troisième polymère) à un polyuréthane à base polycaprolactone.

5 8. Compositions selon l'une quelconque des revendications 1 à 7, caractérisées en ce que ledit troisième polymère, lorsqu'il est présent, l'est dans une proportion de 0 à 20 % en poids par rapport au mélange global.

10 9. Compositions selon l'une quelconque des revendications 1 à 6, caractérisées en ce qu'elles associent au polyuréthane un poly- β -hydroxybutyrate ou un polyhydroxyvalérate.

15 10. Application des compositions selon l'une quelconque des revendications 1 à 9 à l'obtention de feuilles, films ou plaques biodégradables.

20 11. Application des compositions selon l'une quelconque des revendications 1 à 9 à l'obtention de feuilles, films ou plaques biodégradables par calandrage.

12. Feuilles, films ou plaques biodégradables obtenus à partir des compositions selon l'une quelconque des revendications 1 à 9.

25 13. Application des feuilles, films, plaques biodégradables selon la revendication 12 comme enveloppes mortuaires, films agricoles, films d'emballage souples, matériaux de remplissage ou de rembourrage.



INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification ⁵ : A23P	A2	(11) International Publication Number: WO 93/05668 (43) International Publication Date: 1 April 1993 (01.04.93)
(21) International Application Number: PCT/CA92/00406 (22) International Filing Date: 18 September 1992 (18.09.92) (30) Priority data: 761,221 19 September 1991 (19.09.91) US (60) Parent Application or Grant (63) Related by Continuation US 761,221 (CIP) Filed on 19 September 1991 (19.09.91) (71)(72) Applicant and Inventor: AUNG, David [CA/CA]; 7 Kimbercroft Court, Scarborough, Ontario M1S 4K7 (CA).		(74) Agent: BERESKIN & PARR; 40 King Street West, 40th Floor, Toronto, Ontario M5Y 3Y2 (CA). (81) Designated States: AT, AU, BB, BG, BR, CA, CH, CS, DE, DK, ES, FI, GB, HU, JP, KP, KR, LK, LU, MG, MN, MW, NL, NO, PL, RO, RU, SD, SE, US, Euro- pean patent (AT, BE, CH, DE, DK, ES, FR, GB, GR, IE, IT, LU, MC, NL, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, ML, MR, SN, TD, TG). Published <i>Without international search report and to be republished upon receipt of that report.</i>
(54) Title: COMPOSITION SUITABLE FOR FORMING INTO SHAPED ARTICLES, PROCESS FOR PREPARING THE COMPOSITION, PROCESS FOR PREPARING SHAPED ARTICLES USING THE COMPOSITION, AND SHAPED ARTICLES SO-FORMED (57) Abstract A composition suitable for forming into shaped articles comprising flour, starch, and water. The flour, starch and water are each present in an amount such that the composition is rigid and stable over a predetermined temperature range. The composition preferably contains 40 to 80 % by weight flour, 20-60 % starch and 15-25 % water and the flour and starch are obtained from natural cereal sources, such as corn, rice, potato, tapioca and wheat. A process of preparing the composition and a shaped article, and a shaped article produced in accordance with the process are also described. The shaped article may be used for foodstuffs.		

FOR THE PURPOSES OF INFORMATION ONLY

Codes used to identify States party to the PCT on the front pages of pamphlets publishing international applications under the PCT.

AT	Austria	FI	Finland	MN	Mongolia
AU	Australia	FR	France	MR	Mauritania
BB	Barbados	GA	Gabon	MW	Malawi
BE	Belgium	GB	United Kingdom	NL	Netherlands
BF	Burkina Faso	GN	Guinea	NO	Norway
BG	Bulgaria	GR	Greece	NZ	New Zealand
BJ	Benin	HU	Hungary	PL	Poland
BR	Brazil	IE	Ireland	PT	Portugal
CA	Canada	IT	Italy	RO	Romania
CF	Central African Republic	JP	Japan	RU	Russian Federation
CG	Congo	KP	Democratic People's Republic of Korea	SD	Sudan
CH	Switzerland	KR	Republic of Korea	SE	Sweden
CI	Côte d'Ivoire	LI	Liechtenstein	SK	Slovak Republic
CM	Cameroon	LK	Sri Lanka	SN	Senegal
CS	Czechoslovakia	LU	Luxembourg	SU	Soviet Union
CZ	Czech Republic	MC	Monaco	TD	Chad
DE	Germany	MG	Madagascar	TG	Togo
DK	Denmark	ML	Mali	UA	Ukraine
ES	Spain			US	United States of America

- 1 -

Title: COMPOSITION SUITABLE FOR FORMING INTO SHAPED ARTICLES, PROCESS FOR PREPARING THE COMPOSITION, PROCESS FOR PREPARING SHAPED ARTICLES USING THE COMPOSITION, AND SHAPED ARTICLES SO-FORMED

5 FIELD OF THE INVENTION

The invention relates to a composition for forming into shaped articles; a process for preparing the composition; process for preparing shaped articles using the composition; shaped articles so-formed and an
10 apparatus for forming the composition into shaped articles.

BACKGROUND OF THE INVENTION

Disposable packaging is widely used for a variety of products, including both hot and cold
15 convenience foodstuffs and beverages such as hamburgers, french fries, coffee, sandwiches and the like. Disposable packaging is convenient because it is inexpensive, requires no washing and can be discarded after use.

Commercially available disposable packaging
20 suffers from a number of disadvantages, one of the most significant being environmental problems. Such packaging is typically made from oil-based plastics or other artificial materials such as polystyrene foam. These materials are not biodegradable and are disposed of in
25 landfill sites where they accumulate and persist indefinitely as environmental contaminants. Packaging derived from paper products is manufactured using a process that requires the destruction of forest reserves and produces contaminating byproducts. Currently
30 available disposable packaging is also expensive to produce.

Biodegradable containers have been reported for use in packaging foodstuffs. United States Patent No. 3,549,619 to Mark and Mehlretter teaches a method for the
35 preparation of amylose acetate dispersions capable of yielding edible transparent films suitable for packaging of food. The water-soluble food packaging films are

SUBSTITUTE SHEET

produced from high amylose corn starch acetylated with acetic anhydride. The resulting corn starch acetate granules are cooked by steam jets at 177°C to disintegrate the granules. Water-soluble food packaging films are then
5 cast from the resulting amylose acetate aqueous dispersions. The product is a water-soluble, edible, flexible film which is especially suited to package dry foods intended to be added to liquid prior to use, such as coffee or soup.

10 Biodegradable polymers, such as starch, have been incorporated into oil-based plastics. A corn starch-based additive is often used at a concentration of between 6 and 15% of the final product. In the appropriate environment, such as a landfill site, microorganisms
15 digest the starch. Bulk biodegradation occurs at concentrates approaching 50% starch but, as the percentage of starch increases there is a concomitant loss of physical properties of the plastic (See Modern Plastics Encyclopedia Mid-October 1990 issue p. 178).

20 United Kingdom patent application No. 2,029,836, discloses a method and composition of materials for preforming starch with a lubricating fluid into pellets for use in the extrusion of biodegradable plastic. However, the so called biodegradable plastics are
25 comprised of traditional oil-based plastic polymers loaded with starch or other rapidly decomposing material as a binder. The binding material breaks down rapidly but leaves small particles of the plastic polymer which are not biodegradable. The oil-based plastics blended with
30 biodegradable materials such as starch have not gained commercial acceptance because they are not fully biodegradable and they are expensive to produce.

Natural polymers which can be processed by conventional plastics technology have been obtained as
35 fermentation products from single cell microorganisms. Biocycle, March 1989, p. 58 discloses the isolation of a biodegradable polymer poly(3-hydroxybutyrate-3

hydroxyvalerate). However, the polymer is expensive to produce and can not compete commercially with oil-based plastics.

Biodegradable containers which disintegrate when placed in the ground are known for use in plant cultivation and transplantation. These biodegradable plant transplanter containers may be manufactured from polymers such as polylactones or oxyalkanoyl polymers and naturally occurring biodegradable material such as rice hulls, brewers yeast, fir bark or cellulosic products. European Patent Application No. 355,250 discloses a porous container for plant cultivation prepared by mixing rice grain husks with water and a polyurethane prepolymer, molding to the desired shape and air drying.

There is a need for biodegradable packaging material derived from natural products which is environmentally friendly and prepared by a commercially useful process. In particular there is a need for biodegradable packaging suitable for packaging a wide variety of foodstuffs including hot and cold liquids and hot foods with a high fat content, such as hamburgers and french fries.

SUMMARY OF THE INVENTION

The present invention provides a composition suitable for forming into shaped articles comprising flour, starch, and water, said flour, starch and water each being present in an amount such that said composition is rigid and stable over a predetermined temperature range.

Preferably the composition contains 40 to 80% by weight flour, 20-60% by weight starch and 15-25% by weight water and the flour and starch are obtained from natural cereal sources, such as corn, rice, potato, tapioca and wheat. Preferably the particle size of the flour and starch is in the range of about 150-250 mesh and the moisture content is in the range of about 8.0-15% by weight and 10-23% by weight, respectively. The

composition may also contain additives such as softening agents, firming agents, colouring agents, flavouring agents, anti-sticking agents, anti-staling agents and anti-oxidants depending on the desired result to be
5 achieved.

Most preferably the flour, starch and water are each present in an amount in the composition such that the composition has a compression strength of 10 to 60 MPa when measured by a test conducted in accordance with ASTM
10 D-695, an insulating capability of 1.2 to 4.5 cal. cm²/s.cm.c when measured by a test conducted in accordance with ASTM C-177, a thickness of about 80 to 250 mil (about 1.016-5.080 mm) when measured by a test conducted in accordance with ASTM D-1005-84, a moisture content of
15 10-19% by weight when measured by a test conducted in accordance with AACC 44-15A, a degree of expansion of 300-500% when measured by a test conducted in accordance with ASTM D-1005-84, and/or a density of about 0.06-0.8 g/cc when measured by a test conducted in accordance with
20 ASTM D-792-86.

The invention also relates to a process for preparing a composition suitable for forming into shaped articles comprising preparing a mixture of flour and starch wherein the flour and starch have a uniform
25 particle size; and heating and mixing the mixture under a sufficient pressure, temperature and moisture content and for a sufficient period of time such that when the pressure is decreased the mixture expands to form a composition which when cooled is rigid and stable over a
30 predetermined temperature. Preferably the mixture of flour and starch is prepared using 40 to 80% by weight flour and 20-60% by weight starch and the flour and starch are obtained from natural cereal sources, such as corn, rice, potato, tapioca and wheat. Preferably the particle size
35 of the flour and starch is in the range of about 150-250 mesh and the original moisture content of the flour and starch is in the range of about 8.0-15% by weight and 10-

23% by weight, respectively. Additives such as softening agents, firming agents, anti-sticking agents, anti-staling agents, colouring agents, flavouring agents and anti-oxidants may be added at any step in the process depending
5 on the desired result to be achieved.

A shaped article may be formed using the composition of the invention by injecting the composition into a molding press and forming the composition into a shaped article. The shaped article may then be coated with
10 a water repellent coating. Accordingly, the present invention relates to a shaped article produced using the composition of the invention and most preferably a shaped article produced by a process of the invention.

The properties of the composition of the
15 invention make it suitable for making shaped articles to contain foodstuffs. In particular, the composition of the invention may be entirely composed of natural edible ingredients such as flour and starch and accordingly is biodegradable and edible. It will be appreciated that the
20 addition of minor amounts of additives to the composition of the invention will not significantly affect the biodegradable and edible properties of the composition. The composition of the invention is also rigid, light weight, and stable over a temperature range of about -40°C
25 to 160°C, has a compression strength of 10 to 60 MPa when measured by a test conducted in accordance with ASTM D-695 and an insulating capability of 1.2 to 4.5 cal. cm²/s.cm.c when measured by a test conducted in accordance with ASTM C-177, a thickness of about 80 to 250 mil (about
30 1.016-5.080 mm) when measured by a test conducted in accordance with ASTM D-1005-84, a moisture content of 10-19% by weight when measured by a test conducted in accordance with AACC 44-ISA, a degree of expansion of 300-500% when measured by a test conducted in accordance with
35 ASTM D-1005-84, and/or a density of about 0.06-0.8 g/cc when measured by a test conducted in accordance with ASTM D-792-86. The composition of the invention is thus

suitable for direct contact with foods, in particular it would be useful for forming into shaped articles for containing hot water-based liquids. The composition also has a practical shelf life. Further, the invention provides a commercially useful process for preparing the composition and shaped articles of the invention.

The invention also relates to an apparatus for forming a composition comprising flour, starch and water, into a shaped article, the apparatus comprising (1) a cooking device for cooking the composition at an elevated temperature and pressure and having an outlet for the cooked composition; and (2) a die means having an inlet connected to the outlet of the cooking device and defining said shaped article. Preferably, the cooking device of the apparatus comprises a cooker extruder device having an extruder outlet, through which the cooked composition passes. The apparatus may include pump means connected between the outlet of the cooking device and the inlet of the die means for pumping the cooked composition into the die means. The die means may also include means for heating and means for cooling an article formed therein.

BRIEF DESCRIPTION OF THE DRAWINGS

The invention will be better understood with reference to the drawings which show a preferred embodiment of the present invention and in which:

Figure 1 is a schematic drawing showing a process for producing a rigid biodegradable, edible food container;

Figure 2 is a schematic drawing showing a die/press molding system for producing a rigid biodegradable, edible food container;

Figure 3 is a drawing showing the die/press molding apparatus of the invention;

Figure 4 is a schematic side view of another apparatus according to the present invention;

Figure 5 is a section along the axis of a mould according to the present invention;

Figure 6 is an end view of the mould of Figure 5;

Figure 7a and b are an end view and sectional view of an outer part of a slide nozzle; and

5 Figure 8a and b are an end view and a sectional view of an inner, nozzle part of the slide nozzle.

DETAILED DESCRIPTION OF INVENTION

As hereinbefore mentioned the present invention provides a composition suitable for forming into shaped
10 articles comprising flour, starch, and water, said flour, starch and water each being present in an amount such that a shaped article formed from said composition is rigid and stable over a predetermined temperature range.

Preferably the composition contains 40 to 80% by weight
15 flour, 20-60% starch and 15-25% water and the flour and starch are obtained from natural cereal sources, such as corn, rice, potato, tapioca and wheat. A variety of additives may be used in the composition depending on the desired result. Examples of suitable additives are
20 provided in Table I. For further examples of suitable additives see R.S. Igoe, Dictionary of Food Ingredients, Van Nostrand Reinhold, New York, 2nd Ed., 1989; and R.J. Lewis Sr., Food Additives Handbook, Van Nostrand Reinhold, New York, 1989.

25 Most preferably the flour, starch and water are each present in an amount in the composition such that a shaped article formed from the composition has a compression strength of 10 to 60 MPa when measured by a test conducted in accordance with ASTM D-695, an
30 insulating capability of 1.2 to 4.5 cal. cm²/ s.cm.c when measured by a test conducted in accordance with ASTM C-177, a thickness of about 80 to 250 mil (about 1.016-5.080 mm) when measured by a test conducted in accordance with ASTM D-1005-84, a moisture content of 10-19% by weight
35 when measured by a test conducted in accordance with AACC 44-I5A, a degree of expansion of 300-500% when measured by a test conducted in accordance with ASTM D-1005-84, and/or

a density of about 0.06-0.8 g/cc when measured by a test conducted in accordance with ASTM D-792-86.

The composition of the invention may be in pellet form or any other form suitable for employing in a process for preparing shaped articles.

As hereinbefore mentioned, the invention also relates to a process for preparing a composition suitable for forming into shaped articles comprising preparing a mixture of flour and starch wherein the flour and starch have a uniform particle size; and heating and mixing the mixture under a sufficient pressure, temperature and moisture content and for a sufficient period of time such that when the pressure is decreased the mixture expands to form a composition which when cooled is rigid and stable over a predetermined temperature. Preferably the mixture of flour and starch is prepared using 40 to 80% by weight flour and 20-60% by weight starch and the flour and starch are obtained from natural cereal sources, such as corn, rice, potato, tapioca and wheat. Preferably the particle size of the flour and starch is in the range of about 150-250 mesh and the original moisture content of the flour and starch is in the range of about 8.0-15% by weight and 10-23% respectively. Additives such as softening, firming, anti-sticking, and anti-staling agents and anti-oxidants may be added at any step in the process depending on the desired result to be achieved.

As hereinbefore mentioned the composition of the invention may be in pellet form. Preferably, the pellet comprises the elements of the composition in an unexpanded form and the pellet is formed by preparing a mixture of flour and starch wherein the flour and starch have a uniform particle size, heating and mixing the mixture under sufficient pressure, temperature and moisture content and for a sufficient period of time such that the resulting material does not expand when it is extruded from an extrusion cooker. Preferably the moisture content

is 10-12%, the temperature is 240-280°F and the pressure is 50-100 psi. The pellets may be heated and mixed under a sufficient pressure, temperature and moisture content and for a sufficient period of time such that when the pressure is decreased the pellets expand to form a composition which when cooled is rigid and stable over a predetermined temperature.

A shaped article may be formed using the composition of the invention by injecting the composition into a molding press and forming the composition into a shaped article. The shaped article may then be coated with a water repellent coating.

A preferred embodiment for producing shaped articles is shown in Figure 1 and described below.

Feed materials, comprising flour, starch and additives are stored in feed silos 1-3. The flour and starch are obtained from natural cereal sources, such as corn, rice, potato, tapioca and wheat. The particle size of the flour and starch are preferably in the range of 150-250 mesh and the moisture content is in the range of 8.0-15% by weight and 10-23% respectively as shown in Table II. The starch may be obtained from C. Tennant, Canada and Maize, U.S. (corn), Rhone-Poulenc (potato); A. Stanley, U.S. and Nacan, Canada (tapioca); and Manildra, U.S. and Prescott, Canada (wheat) the flour may be obtained from J. Short, U.S.; Dover, Canada (corn); Comet, U.S. Grain Products, Canada (rice) and N. Dakota, U.S. and Grain Products, Canada (wheat). As discussed above, a variety of additives may be used depending on the nature of the shaped article to be produced.

The feed materials are fed into weigh mixer 4 in the following proportions, 40-80% flour, 20-60% starch, 15 to 25% water and 0-10% additives. The mixture is sifted in a sifter 5 to provide particles of a uniform size and passed through a metal detector 6 to detect and remove any metal which may have inadvertently been incorporated into the mixture. The mixture is held in storage silo 7 before

passing to a volumetric feeder 8. Water, flavouring and colouring agents are held in storage tanks 9, 10 and 11 respectively.

The flavouring agent may be one or more of a natural flavour or an artificial flavour or an artificial or a combination of natural and artificial flavours. Examples of suitable natural flavouring agents include grapefruit oil, jasmine oil, lemon oil, lime oil, orange oil and rose oil. The desired result as well as the nature of the flavouring agent will determine the actual amount used in any particular incident.

The colouring agent may be a natural or artificial colouring agent or a combination of both. The amount of colouring agent to be added can be determined by visual requirement. Natural colouring agents such as saffron, paprika, beetroot, crocein and carotene are preferably used as colouring agents.

Flavouring and colouring agents are most preferably selected from the relevant list of approved agents, for example those approved by Health and Welfare, Canada and the Federal Drug Agency, United States.

In the cooking phase the sifted mixture, water and any colouring and flavouring agents are fed into extrusion cooker 12 at a feed rate of 75-100 lb/hour and a nozzle ratio of 15:1 to 25:1 with a single or twin screw rotating at a speed of 100-350 rpm, preferably the extrusion cooker is an MPF 50/35 extrusion cooker or an MPF 40D extrusion cooker obtained from APV Baker Inc. with a co-rotating twin screw. The dough is mixed, kneaded and cooked with a moisture content of 15-35% at a temperature of 120-280°F and a pressure of 200-300 psi. The pressure is reduced by venting at the outlet of the cooker 12, resulting in expansion of the dough. In known manner, release of the pressure causes the water present to instantaneously vaporise, causing the puffing on expansion. The expanded hot dough is pressure injected.

into molding press 13, which has a water-cooled die mold, as detailed below. The expanded hot dough fast cools in the molding press 13 at the surface of the cold die mold. The expanded packaging material is molded to the correct
5 thickness in the molding press 13.

The formed packaging material may then be coated with a water repellent, such as Methocell® in a coater 14, (for example, a water resistance coating machine manufactured by Christy Machine Company, model: tube/cone
10 coater; powder and liquid dispenser) dried in an oven 15 and cooled in a cooling chamber 16 (for example, a cooling/cutting machine model: Multi-zone, manufactured by Greerco). The finished product can then be processed through stacking and packing machines 17 and 18 (for
15 example, the stacking and packing machine model Dyna-Pak manufactured by Eagle Packing) prior to shipping. A heat exchanger 19 can be incorporated into the system to conserve energy as shown in Figure 1.

Instead of being formed directly into a usable
20 packaging article the sifted mixture may be formed into pellets by feeding the mixture into the extrusion cooker 12. The operating conditions of the extrusion cooker are selected such that no puffing or expansion of the extrudant takes place in the extrusion cooker. Preferably
25 the moisture content is 10-12%, the temperature is 240-280°F and the pressure is 50-100 psi, i.e. the pressure, temperature and moisture content are selected to prevent any significant puffing or expansion occurring. The extrudant is then cut into pellets preferably by blades at
30 the end of the cooking extruder, and the pellets are dried and cooled to room temperature. The pellets may then be used in the process described above in place of the sifted mixture.

It is expected that the above described
35 technique to produce pellets would be useful for a principal supplier who can ensure that the pellets are manufactured to the correct formulation. The pellets

- 12 -

could then be supplied to secondary suppliers who would process them as described to produce the finished packaging articles. This can also save on transportation costs. Each secondary supplier can be located close to
5 purchasers of the packaging articles, and the principal supplier would then only have to distribute the compact pellets which could be shipped in bulk.

The steps in the extrusion molding process may be better understood by reference to Figure 2 which is a
10 schematic drawing of the die/press molding system. Figure 3 is a detailed drawing of the apparatus. The ingredients can either be supplied by the volumetric feeder 8 and tanks 9, 10 and 11, or as pellets from a storage hopper (not shown). In the later case, water may be added to
15 adjust the moisture content, and the flavouring and colour content could also be adjusted at this time by addition of further flavouring and/or colouring agents. In the extrusion cooker 12 the ingredients are mixed, cooked and expanded to form a puffed extrudant. The screw rotational
20 motion and cooker internal pressure push the extrudant into the next stage. Upon exiting the cooker/extruder 12, the water present, due to its elevated temperature and pressure, vaporises. This simultaneously causes the mixture to expand or puff, and also to cool.

25 In the next stage, the die adaptor 20 having an inlet 21 and injector 22, receive the puffed extrudant (temperature of 258-287°F; moisture content of 10-20%, an expansion factor of 400-500%, and a pressure of 3,000 to 35,000 psi). The extrudant is injected/pumped by means of
30 external pressure system 24 into the molding press 26. The pressure system 24 includes pistons 24a, a hydraulic/pneumatic inlet 23, and an outlet 25 to a compressor. Provision for cooling by a suitable coolant is made at 29. The external pressure system 24 is
35 preferably a high pressure pump with a piston such as the dual piston pump manufactured by Cole-Parmer, (100 rpm, 5,000-12,000 psi, 20-200°F). As relatively high pressures

are used, sufficient puffing or expansion occurs through the vaporisation of water, and no other expansion agent is required. The molding press 26 defines a die cavity 27.

In the molding press 26 the expanded extrudant
5 is formed, in the die cavity 27, into the desired three dimensional shape of the container 28 and the interior surface of the container is coated, with cold or hot die/molding compression as more particularly described below. A steam or electric heating unit 30 and a water
10 cooling unit 32 provide the heat or cooling required. The cooling unit 32 has an inlet 32a for cold water and an outlet 32b for warmed water. The cold die is maintained at a temperature of 20-40°C and the hot die at a temperature of 100-160°C. The molding press 26 is operated
15 under a pressure of from 5,000-30,000 psi. In particular, a coating material, preferably a water repellent may be incorporated into the starting ingredients and a coating may be formed on the interior surface of the container 28 by heating at a temperature of 100-160°C using the steam
20 or electric heating unit 30 under a pressure of 5,000-30,000 psi. The die is then cooled using the water cooling unit 32.

Examples of suitable water repellents include cellulose such as ethyl cellulose; proteins, such as
25 casein, gluten, glutenin, and zein; and alginates such as sodium and calcium alginate and in particular water repellents sold under the trade marks Algin (Prescott Company), Ethocel (Dow Chemicals), Prolait (Charles Tennant), Modglut (Ogilvie Mill), and Zein (Freeman
30 Industries). The water repellent, preferably casein, gluten or zein, may be incorporated into the starting ingredients and a water repellent coating may be formed on the interior surface of the container 28 as described above. Alternatively, the water repellent may be spray
35 coated onto the formed container by powder or liquid spray coating methods using conventional techniques (see for example Finishing Guidebook 1988, Metal Finishing

Magazine; Deposition Technologies for Film and Coatings, Roinstant F. Banshah; Metal Handbook Vol. 2, American Society of Metal; Chemical Engineer's Handbook, Robert H. Perry). Preferably water repellents such as Ethocel® and
5 alginate are employed in the latter method.

The shaped articles produced in accordance with the process of the invention are sufficiently rigid, impermeable and water repellent to hold moist or fatty whole foods and liquids, including cold or hot beverages.
10 The material which forms the walls of the shaped articles has a light and expanded or puffed texture, similar in nature (e.g. self-skinned and closed celled) to that of the polystyrene foam commonly employed in disposable coffee cups and food trays. Unlike polystyrene products,
15 the containers of the present invention may be made from natural products and are completely biodegradable and edible. The puffed nature of the shaped articles of the invention provides a light weight product which can be used to insulate hot or cold foods and beverages, such as
20 coffee, hamburgers, ice cream and cold drinks. The shaped articles can take a wide variety of forms, including, but not limited to, containers, boxes, cups, lids, plates, trays, straws, eating utensils and structure blocks. The following examples are illustrative of the present
25 invention:

Example 1

A range of formulations as shown in Table III were tested using the above described process and the conditions set forth in Table VI, employing the eight
30 basic ingredients of corn flour, potato flour, rice flour, wheat flour, corn starch, potato starch, tapioca starch and wheat starch, to determine the preferred formulations for preparing rigid packaging material. The preferred formulations are shown in Table IV.

35 The particular formulations shown in Table V were processed as generally described above to obtain a mixture with particles having a mesh size of 200 and a

moisture content of 11% by weight. A dry feeder was used to meter each formulation and the extruder and screw configuration were set up to include a mixing zone for the raw materials. The ingredients were cooked in a MPG 50/35
5 extrusion cooker (APV Baker Inc.) with a co-rotating twin screw using the conditions set forth in Table VI with the extruder screw in configuration 2 as detailed in Table VI. Preferably, the procedure was carried out under a pressure of 250 psi, moisture content of 22.5%, feed rate of 100
10 lb/hour for 35 seconds. A total of 6 zones of extruder-barrel were utilized and each zone temperature was set up to cook/expand in the extruder. The screw ratio was 15:1 and the temperatures increased from 80°F in zone 1 to 280°F in zone 6. Water was injected into the mixing zone of the
15 extruder with a water pump. The degree of expansion of the extrudant depended on the amount of water infused.

The cooled expanded extrudant was fed into the die adaptor and injector (dual piston pump manufactured by Cole-Parmer, 100 rpm, 5,000-12,000 psi) at a temperature of
20 about 260°F, a moisture consistent of 10-20% and an expansion factor of 400-500%. The adaptor and injector was powered either by a hydraulic or by a pneumatic compression system maintained at a temperature of 258-287°F and a pressure of 5,000 psi. The adaptor and
25 injector injected the expanded extrudant into the molding, or die/form press. The die/form press was maintained at a pressure of 12,000 psi. The three dimensional shaped containers were formed in the die/form press, which was steam heated to 310°F and water cooled to 80°F in alternate
30 cycles. The heat and pressure in the die/form press formed the water repellent coating (about 20 ml thickness) in these formulations which incorporated casein, zein, or gluten as the coating material.

In an alternative procedure the water repellent
35 was coated on the pre-molded product. In this case Ethocel® or alginate was coated on the product by powder or liquid spray coating methods (nozzle press - 100 psi).

The formed containers were dried in an electrical or gas powered oven for 3.5 minutes at 280°F.

The containers were tested for moisture content (AACC 44-15A, Sartorius MA 30), thickness/expansion degree (ASTM D-1005 -84, Fowler micrometer), insulation (ASTM C-177-85, Thermal conductivity Tester manufactured by Holometrix, type K-Matic/Rapid K, 0.5-100), compression strength (ASTM D-695-89, Chatillon/BG 100), density (ASTM D-792-86, Satorius B120 S) and water resistance (ASTM D-870-87, Precision Bath/tank manufactured by Cole-Parmer, Digital type, 68-302°F, 14 litre). The stability of the containers was tested using an oven heating technique. The containers were placed in an oven, the temperature was increased in 5°F increments from room temperature, and the temperature at which decolorization and cracking occurred was recorded. The colour, odour and flavour of the materials was also noted. The results are shown in Table VI.

An aluminum plate/tray, a paper cup (Canada Cup, Lily Cup, Canada) a paper plate/tray (CKF), a plastic cup (Canada Cup, Lily Cup, Canada) and a plastic plate/tray were tested for thickness, compression strength, insulation strength, density, water resistance, and stability using the same methods as used for the test formulations. The results are shown in Table VIII. A comparison of the results shown in Table VI and VIII indicates that the properties of the tested formulations were similar to the properties of the plastic materials tested.

Reference will now be made to Figures 4 - 8, which show an alternative embodiment of the invention.

In Figure 4, there is shown a platform 40 for supporting apparatus. Above this is supported a mixer 42, which is connected to a volumetric feeder 44. The volumetric feeder 44 in turn supplies a hopper 46. A control panel 48 includes the necessary control instrumentation devices.

An extruder 50 has a barrel and screw in known manner, and is driven by a motor 52. The extruder 50 is a single screw extruder, and here is an Engel Model ES55 Injection Molding Machine, as produced by Engel Canada
5 Inc.

The screw and barrel configuration should be chosen to give desired characteristics. Here, these are: mixing; cooking; pressurising; and expansion. To ensure adequate mixing, a modified barrel profile was used
10 including channels.

The end of the extruder barrel is connected to a mold 54 comprising a mold cavity 55 and a mold core 56. The mold core 56 is mounted to a support plate 58, which in known manner is mounted for sliding movement on shafts
15 60, for engagement and disengagement with the cavity 55.

Referring to Figures 5 and 6, these show in greater detail the mold 54. The mold cavity 55 defines an internal cavity 62 having the shape of a coffee or other beverage cup. The shape of the cup can be generally
20 conventional, and details of its shape form no part of the present invention. The base of the cup, indicated at 64, has a lip 66 around its edge. In the middle of the base 64, a duct opens for supplying the material into the cavity 62.

The core 56 comprises a core body 68, mounted on the support plate 58 and a stripper plate 72. The stripper plate 72 can be displaced away from the plate 58, when the mold is open, to displace or strip a finished cup from the core 56. As indicated at 74, there is a gap of
30 0.004 inches between one end of the core body 68 and the stripper plate 72. This is to provide venting of air as material enters the mold cavity 62. An inlet plate 70 also provides a gap of four thousandths of an inch, as indicated at 71. Both gaps or vents 71, 74 are vented to
35 the exterior with the gap 74 being vented via annular channel 75.

A central bore 76, with a closed end, is

provided for a heating element, and a smaller, offset bore 78, also closed ended, is provided for a thermocouple or other temperature measuring device. As shown in Figure 6, the bore 76 extends into an open channel 77 for electrical connections. Three cylindrical bores 80 are symmetrically arranged around the edge of the mold 54, for mounting on the shafts 60. Alternating with the bore 80 are three other bores 81, having a smaller diameter through the stripper plate 72 and part of the mold cavity 55. These are for shoulder bolts, which permit limited movement of the stripper plate in known manner.

Turning to Figures 7 and 8, these show details of the sliding nozzle seal. This comprises a first nozzle part or nozzle body 82, which is threaded at one end, as indicated at 84, for securing in the end of the barrel body 50. The nozzle body 82 defines a bore 86. As shown in Figure 7, this bore 86 comprises a relatively wide inlet portion 86a with a diameter of 0.75 inches, an immediate portion 86b of slightly narrower diameter, and an upper or outlet end portion 86c of yet narrower diameter. The portion 86c has a diameter 0.5 inches. The overall bore 86 and nozzle body 82 have a length of 3.25 inches.

The nozzle or second part of the nozzle assembly is shown at 88 in Figure 8, and comprises an upper or outlet portion 90, having a hemispherical abutment surface 92 for abutting a corresponding surface of the inlet plate 70. A bore 94 extends from the hemispherical surface 92 through to a short cross bore 95.

The inlet part 88 has an inlet end portion 96, into which the bore 94 extends. At the lower or inlet end of the inlet portion 96, a screw 98 secures an end member 100. This end member 100 has a diameter of 0.74 inches, and hence is sized to fit within the bore part 86 with a clearance of 0.010 inches. In use, the nozzle part 88 is slidably mounted in the bore 86, and the end member 100 retains the two elements together.

Although not shown, the inlet plate 70 is provided with a hemispherical recess, corresponding to the end surface 92, and a short bore leading from this into the cavity 62.

5 To inject material into the mold cavity 62 and mold a cup, in known manner, the mold core 56 and cavity 55, together with stripper plate 72 are sandwiched between the plate 58 and barrel 50. This causes the nozzle part 88 to be pressed downwards within the nozzle body 82, as
10 viewed in Figures 7 and 8, bringing the end portion 90 into abutment with the nozzle body 82. The lower face of the cylindrical member 100 is spaced by 2.938 inches from an abutment face 102 of the end portion 90. As this is less than the overall length of the nozzle body 82, this
15 ensures the end member 100 is retained in the bore part 86a and cannot extend beyond it. A spacing of 2.688 inches between the abutment face 102 and the end member 100 enables the nozzle part 88 to reciprocate in the two bore portions 86b, c between a closed position and an
20 injection position.

In the closed position, the nozzle member 100 abuts an annular stop between the bore portions 86b, c, and the cross bore 95 is within the bore portion 86c, and hence effectively is closed off. The bore portion 86c and
25 the nozzle end portion 96 have closely similar diameters for this purpose. In the injection position, the abutment surface 102 abuts the top of the nozzle body 82, and the cross bore 95 is located in the bore portion 86b. This enables material to be injected around the end element
30 100, through the bore parts 86b and c, through the cross bore 95 and bore 94, and through the bore of the inlet plate 70 into the mold cavity 62.

After a predetermined amount of the material has been injected into the mold cavity 62, the injection is
35 stopped, and the pressure applied by the barrel 50 released. This permits the nozzle part 88 to travel to the closed position, under the influence of the pressure

within the nozzle body 82. The cross bore 95 is then located in the narrow bore portion 86c, and hence closed. At this time, the mold is opened, with the plate 58 drawing the mold core 56 out of the mold body 55. The
5 stripper plate 72 is permitted to travel some distance, but the operation of the shoulder bolts is such as to cause it to strip the formed cup from the mold core 56, in known manner.

To facilitate separation of the various
10 components, and prevent the material used from sticking to the various components, a Teflon coating is used. All surfaces forming the mold cavity were coated with Teflon. Additionally, the stripper plate 72 and surfaces defining the vent conduits at 74, 75 were also coated with Teflon.
15 Additional, the inlet plate 70 and surfaces defining the vent 71 were coated with Teflon. Coating the inlet plate 70 with Teflon also facilitated separation from the sliding nozzle 82, 88.

A number of tests were carried out using the
20 apparatus described above. These were carried out using a mixture comprising one hundred parts dry material, selected from the formulations given in Table V, two parts per hundred of MAGIC baking soda, and twenty-seven parts per hundred of water. The dry ingredients were thoroughly
25 mixed before water was added.

As the equipment was designed for free-flowing material, and as the mixture was not sufficiently free-flowing to prevent bridging in the mixer 42 and/or feeder 44, the material was hand fed to the throat of the barrel.
30 Starve feeding was used, since it was found that otherwise inconsistent sprue feed times were obtained. Starve feeding involves feeding just sufficient material to the barrel for each cycle. In this case, 24 grams of material was sufficient to give a uniform screw feed time.

35 Mold surface temperature was between 275°F - 330°F. When the temperature increased to 373°F for the core and 344°F for the cavity, too much blowing occurred,

and material was extruding out of the sprue. With temperatures then decreased to core 305°F and cavity 310°F, satisfactory results were obtained.

In initial testing, while the cups were generally satisfactorily formed, parts of the cup were not completely formed; for example, part of the bottom lip was missing on occasion. This used an old design with a lesser degree of venting. With the mold design described above, with vents 71, 74 having a four thousands of an inch width, it was found there was sufficient venting for the cups to be formed consistently.

It was found that the material continued to extrude out the back of the sprue in the inlet plate 70 and to expand, riveting the sprue into the mold. This was a continuing problem, and it is believed that the sprue area should not be depressurized until the sprue has "frozen off" or the pressure in the cavity has been dissipated

It was found that the temperature of the nozzle and barrel affected the temperature of the final material. At 300°F, the material was quite dark. At 250°F, a consistent, lighter colour was produced. If too high a temperature was present at the mold 54, then this would cause blowing and the presence of large bubbles in the finished articles. The hot mold has the advantage of giving a high gloss, shiny finish to the finished article, whereas too cold a mold gives a rough finish. It was found that a mold temperature equal to the barrel temperature gave satisfactory results.

From the result of tests carried out, optimum temperatures, pressures and screw rotation speed were found, and these are set out in Table IX. This Table gives, for each parameter, the set range of values, actual values and best values.

As shown, in the feeding section of the barrel, the temperature should be around 82 - 83°F, with the mixing and kneading section of the barrel at 104 - 106°F. The

cooking and expanding sections of the barrel should, respectively, preferably be in the ranges 147 - 154°F and 250 - 252°F. The nozzle, i.e. the sliding nozzle 82 - 88, should be at 250 - 251°F. For this purpose, it can be
5 provided with suitable heating. The mold should generally be maintained at a temperature of 300 - 310°F.

It was found that the best injection pressure should be 750 - 850 psi, with a screw rotation speed 180 - 200.

10 It will be appreciated that the exact pressure and screw rotation parameters will vary depending upon the machine used, and the nature of the article being molded. Whether or not any blowing or expansion agent, such as baking soda, is required will depend upon the injection
15 pressure. In general, if the extruder can generate high pressures so that sufficient expansion is obtained through vaporization of water, then little or no expansion agent may be required. On the other hand, at lower pressures, it is found that an expansion agent was required.

20 Additives and compositions as shown in Table I - V can be used in the apparatus of Figures 4 - 8. Also, as for other examples, a suitable water repellent can either be added to the mixture or applied to the finished article.

25 The properties of articles made on the apparatus were similar to those of the other examples, with regard to the properties listed in Table VII.

30 The present invention has been described in detail and with particular reference to the preferred embodiments; however, it will be understood by one having ordinary skill in the art that changes can be made thereto without departing from the spirit and scope thereof.

Legend to Figure 3

- No. 1 ... Hydraulic/Pneumatic Pressure Inlet
- No. 2 ... Outlet to Compressor
- No. 3 ... Piston
- 5 No. 4 ... Die-Cavity
- No. 5 ... Cooled Water Inlet
- No. 6 ... Warmed Water Outlet
- No. 7 ... Die Adapter/Injector
- No. 8 ... Die/Mould Press
- 10 No. 9 ... Cup
- No. 10 ... Extrudant Inlet form Cooking Extruder
- No. 11 ... Coolant
- No. 12 ... Heating Medium

TABLE I

	Function of Additive	Suitable Additives
5	Softening	Syrup, sugar, sorbitol, glycerol, glucose, sodium lactate, mannitol & others
	Firming agent	Aluminium & calcium sulphate, calcium phosphate, chloride, gluconate & others
10	Anti-sticking agent	Silicate of sodium, calcium, calcium-aluminum, magnesium oxide & others
	anti-staling agent	Sucrose, sucrose acetate isobutyrate & others
15	Anti-oxidant	Citric & benzoic acid derivatives, sorbic acid & sorbic sodium, calcium, potassium salt, ascorbic acid & ascorbic acid sodium, calcium salt & others
20	Expansion agent	Sodium bicarbonate, ascorbic acid, sodium lactate

- 25 -

TABLE IITYPE OF FLOUR AND STARCH

<u>TYPE</u>	<u>GRADE</u>	<u>SIZE</u>	<u>MOISTURE</u>	<u>pH</u>	<u>COLOR</u>	<u>FLAVOR</u>	<u>ODOR</u>
<u>FLOUR</u>							
Corn	A	150	11.0-13.0	4.5 / 6.0	white	good	good
Potato	A	150	8.0-10.0	5.0 / 7.0	white	good	good
Rice	No.1	200	8.5-13.0	5.0 / 7.0	white	good	good
Tapioca	No.1	150	10.0-14.0	5.0 / 6.5	white	good	good
Wheat	A	150	13.8-14.2	5.0 / 7.0	white	good	good
<u>STARCH</u>							
Corn	A	200	11.0-12.0	4.5 / 5.5	white	good	good
Potato	A	150	10.0-21.0	5.0 / 7.0	white	good	good
Tapioca	No.1	150	10.0-14.0	5.0 / 6.5	white	good	good
Wheat	A	250	11.0-12.0	5.0 / 7.0	white	good	good

SUBSTITUTE SHEET.

- 26 -

TABLE III

**TESTING FORMULATION FOR RIGID MATERIALS
(PERCENT BY WEIGHT)**

FLOUR	STARCH			ADDITIVES	WATER
	CORN	TAPIOCA	WHEAT		

<u>CORN</u>					
40-100	20-60			0-10	15-25
40-100		20-60		0-10	15-25
40-100			20-60	0-10	15-25
<u>POTATO</u>					
40-100	20-60			0-10	15-25
40-100		20-60		0-10	15-25
40-100			20-60	0-10	15-25
<u>RICE</u>					
40-100	20-60			0-10	15-25
40-100		20-60		0-10	15-25
40-100			20-60	0-10	15-25
<u>WHEAT</u>					
40-100	20-60			0-10	15-25
40-100		20-60		0-10	15-25
40-100			20-60	0-10	15-25

SUBSTITUTE SHEET

TABLE IV

(PERCENT BY WEIGHT)

FORMULATION NO. I

Wheat flour	75 - 85 %
Corn starch	14 - 24 %
Additives	01 - 10 %
Moisture	20 - 23 %

FORMULATION NO. II

Wheat flour	75 - 85 %
Wheat starch	14 - 24 %
Additives	01 - 10 %
Moisture	20 - 23 %

FORMULATION NO. III

Corn flour	75 - 85 %
Corn starch	14 - 24 %
Additives	01 - 10 %
Moisture	20 - 23 %

FORMULATION NO. IV

Corn flour	75 - 85 %
Wheat starch	14 - 24 %
Additives	01 - 10 %

- 28 -

TABLE V

RIGID BIODEGRADABLE/EDIBLE MATERIAL FORMULATIONS (IDEAL)

<u>FORMULATION A</u>		<u>FORMULATION B</u>	
Wheat Flour	.. 74%	Wheat Flour	.. 75%
Wheat Gluten	.. 1%	Wheat Gluten	.. 2%
Corn Starch	.. 20%	Wheat Starch	.. 22%
Sorbitol	.. 2%	Mannitol	.. 1%
Sucrose	.. 1%	Sucrose	.. 1%
Sod Ascorbate	.. 1%	Cal Sorbate	.. 1%
Moisture	.. 23%	Moisture	.. 22%

<u>FORMULATION C</u>		<u>FORMULATION D</u>	
Corn Flour	.. 70%	Corn Flour	.. 71%
Corn Starch	.. 24%	Wheat Starch	.. 25%
Casein	.. 2%	Zein	.. 1%
Glycerol	.. 2%	Sod Lactate	.. 1%
Sod Benzoate	.. 2%	Cal Ascorbate	.. 1%
Moisture	.. 20%	Moisture	.. 22%

SUBSTITUTE SHEET

TABLE VITECHNICAL INFORMATIONBarrel

Ratio	15:1
Moisture	20 - 23 %
Pressure	200 - 300 psi
Temperature	80 - 280 F

Zone	1	2	3	4	5	6
Set (F)	80	80	160	180	240	280
Actual (F)	80	77	161	178	240	280
Residence time (sec)	5	5	5	5	5	5

Screw

Speed	150	-350 rpm
Configuration	#1	- #3
Configuration	#1	#2	#3
Spacer	10D	10D	10D
Bearing	2D	2D	2D
Spacer tube	8D	8D	8D
Feed Screw	4.5D	4.5D	4.5D
Forwarding	7x30	7x30	7x30
Single lead	2D	1D	1D
For	-	3x60	3x60
Rev	3x30	3x30	3x30
Single lead	1.5D	1D	1D
For	2x60	3x60	3x60
Rev	2x30	3x30	3x30
Single lead	1D	1D	1D
For	-	3x60	3x60
Rev	-	3x30	3x30
Single lead	-	1D	1D
For	-	3x60	3x60
Rev	-	3x30	3x30
Single lead	-	1D	1D
For	-	-	3x60
Rev	-	-	3x30
Single lead	-	-	1D

TABLE VII

RIGID BIODEGRADABLE?EDIBLE PACKAGING MATERIAL TEST DATA

TEST NO.	MOISTURE	THICKNESS	EXPANSION DEGREE	COMPRESSION STRENGTH	INSULATION STRENGTH	DENSITY	WATER RESISTANT	COLOR	ODOR	FLAVOUR	STABILITY
I-A	15.35 %	236 mil	400 %	50 MPa	3.5	0.4532 g/cc	good	white	neutral	neutral	326F
II-A	10.69	250	300	46	4.1	0.7724	good	white	neutral	neutral	321
III-A	16.37	160	400	52	2.8	0.3550	good	white	neutral	neutral	325
I-B	15.76	167	300	45	3.6	0.2069	good	white	neutral	neutral	321
II-B	10.30	117	300	37	2.0	0.5509	good	white	neutral	neutral	320
III-B	11.54	195	400	41	2.9	0.5061	good	white	neutral	neutral	338
I-C	12.57	126	400	49	2.5	0.3332	good	white	neutral	neutral	340
II-C	16.82	93	400	53	2.1	0.1037	good	white	neutral	neutral	318
III-C	12.40	144	500	50	4.0	0.0991	good	white	neutral	neutral	336
I-D	18.65	81	350	58	2.6	0.5213	good	white	neutral	neutral	317
II-D	13.98	102	450	43	3.5	0.0762	good	white	neutral	neutral	320
III-D	16.98	139	400	33	4.6	0.0633	good	white	neutral	neutral	310

TABLE VIII
COMPATIBLE RIGID MATERIAL AGAINST
RIGID BIODEGRADABLE/EDIBLE PACKAGING MATERIAL TEST DATA

A = Aluminium ; P = Paper ; Pl = Plastic ; C = Cup ; P/T = Plate/Tray

[illegible]

- 32 -

TABLE IX
PROCESS PARAMETERS (RIGID)

		Temperature (°F)		
		<u>Set</u>	<u>Actual</u>	<u>Best</u>
5	Mould (forming)	280-360	280-360	300-310
	Nozzle 1 (adaptor)	250-300	250-300	250-251
10	Barrel 2 (expanding)	250-300	250-307	250-252
	Barrel 3 (cooking)	32-40	147-165	147-154
	Barrel 4 (mix/kneading)	32-40	93-106	104-106
15	Barrel 5 (feeding)	32-40	77-84	82-83
<u>Pressure and Screw Speed</u>				
20	Injection Press (psi)	750-850	729-1083	750-850
	Screw rotation (rpm)	100-200	109-201	180-200

- 33 -

I CLAIM:

1. A composition suitable for forming into shaped articles by application of elevated temperatures and pressures and subsequent application of reduced pressure
5 to permit expansion of the composition in a mold to form a shaped article, the composition comprising flour, starch, and water, said flour, starch and water each being present in an amount such that said composition is rigid and stable over a predetermined temperature range.
- 10 2. The composition as claimed in claim 1 which contains 40 to 80% by weight flour, 20-60% starch and 15-25% water.
3. The composition as claimed in claim 1 or 2 wherein flour and starch are obtained from natural cereal
15 sources, such as corn, rice, potato, tapioca and wheat.
4. The composition as claimed in claim 1 which further comprises additives selected from the group consisting of softening agents, firming agents, non-sticking agents, preservatives, colouring agents,
20 flavouring agents, expansion agents and anti-oxidant agents.
5. The composition as claimed in claim 1, wherein the flour, starch and water are each present in an amount in the composition such that a shaped article formed from
25 said composition has a compression strength of 10 to 60 MPa when measured by a test conducted in accordance with ASTM D-695, an insulating capability of 1.2 to 4.5 cal. cm²/ s.cm.c when measured by a test conducted in accordance with ASTM C-177, a thickness of about 80 to 250
30 mil (about 1.016-5.080 mm) when measured by a test conducted in accordance with ASTM D-1005-84, a moisture content of 10-19% by weight when measured by a test

- 34 -

conducted in accordance with AACC 44-15A, a degree of expansion of 300-500% when measured by a test conducted in accordance with ASTM D-1005-84, and/or a density of about 0.06-0.8 g/cc when measured by a test conducted in
5 accordance with ASTM D-792-86.

6. A composition as claimed in claim 1 which is in pellet form.

7. A process for preparing a mixture suitable for forming into shaped articles comprising preparing a
10 mixture of flour and starch wherein the flour and starch have a uniform particle size; and heating and mixing the mixture under a sufficient pressure, temperature and moisture content and for a sufficient period of time such that when the pressure is decreased the mixture expands to
15 form a composition which when cooled is rigid and stable over a predetermined temperature range.

8. A process as claimed in claim 6, wherein the flour and starch mixture is prepared using 40 to 80% by weight flour and 20-60% by weight starch.

20 9. A process as claimed in claim 7, wherein the flour and starch are obtained from natural cereal sources, selected from the group consisting of corn, rice, potato, tapioca and wheat.

10. A process as claimed in claim 8, wherein the
25 particle size of the flour and starch is in the range of about 150-250 mesh and the original moisture content of the flour and starch is about 8.0-15% by weight and 10-23% by weight, respectively.

11. A process as claimed in claim 7, 8, 9 or 10,
30 which further comprises adding an additive selected from the group consisting of softening agents, firming agents,

- 35 -

non-sticking agents, preservatives, colouring agents, flavouring agents, expansion agents and anti-oxidant agent.

12. A process as claimed in claim 7, 8, 9, 10 or 11,
5 wherein the mixture is heated and mixed in an extrusion cooker under a moisture content of 15-35%, a temperature of 120-200°C and a pressure of 200-300 psi.

13. A process as claimed in claim 12, wherein the
10 extrusion cooker injects the composition into a mold which defines the shape of the article, and in which the pressure is decreased to permit the mixture to expand to fill the mold.

14. A process as claimed in claim 13, wherein the
15 composition is injected by the extrusion cooker into a mold at a pressure in the range 729 - 1,083 p.s.i.

15. A process as claimed in claim 14, wherein the
20 extrusion cooker: mixes and kneads the mixture at a temperature in the range 93 - 106°F; cooks the mixture at a temperature in the range 147 - 165°F; expands the mixture at a temperature in the range 250 - 307°F, and discharges the mixture at temperature in the range 250-300°F; and wherein the mold is maintained at a temperature in the range 280 - 360°F.

16. A process as claimed in claim 15, wherein the
25 extrusion cooker: is fed the mixture at a temperature in the range 82 - 83°F; mixes and kneads the mixture at a temperature in the range 104 - 106°F; cooks the mixture at a temperature in the range 147 - 154°F; expands the mixture at temperature in the range 250 - 252°F; injects the
30 mixture into the mold at a temperature in the range 250 - 251°F and a pressure in the range 750 - 850 p.s.i.; and wherein the mold is maintained at a temperature in the

- 36 -

range 300 - 310°F.

17. A process as claimed in any one of claims 7 - 16, which includes one of: coating the finished article with a water repellent; and include a water repellent in
5 the mixture.

18. A process as claimed in any one of claims 7 - 16 which further comprises forming a water repellent coating on at least the interior surface of said shaped article by subjecting the interior surface of said shaped article to
10 a temperature in the range 100-160°C.

19. A process as claimed in claim 17, wherein a water repellent agent is coated on said shaped article by powder or liquid spray coating.

20. A shaped article produced in accordance with the
15 process as claimed in any one claim 7 - 19.

21. A shaped article as claimed in claim 20 in the form of a box, cup, lid, plate, tray, bowl or straw.

22. Pellets formed by preparing a mixture of flour and starch wherein the flour and starch have a uniform
20 particle size, heating and mixing the mixture under sufficient pressure, temperature and moisture content in an extrusion cooker, and extruding the resulting material from the extrusion cooker at a temperature and pressure low enough that the mixture does not expand upon
25 extrusion.

23. Pellets as claimed in claim 19, wherein the moisture content is 10-12%, the temperature is 240-280°F and the pressure is 50-100 psi.

- 37 -

24. Pellets as claimed in claim 23, which comprise 40 - 80% by weight flour, 20 - 60% by weight starch and 15 - 25% water.

25. A mold, for molding an article from a composition comprising flour, starch and water, the mold comprising a mold body and a mold core, both of which are coated with a non-stick coating.

26. A mold as claimed in claim 25, which includes at least one vent for air from the mold, and an inlet bore, wherein the vent and the inlet bore are coated with the non-stick coating.

27. A mold as claimed in claim 26, wherein the inlet bore is provided in an inlet plate, and a surface of which is shaped to engage an injection nozzle, wherein the inlet plate is coated with a non-stick coating and defines a vent between the inlet plate and the mold body.

28. A mold as claimed in claim 27, wherein the mold core includes a bore for a heating element and a bore for a temperature sensing device, both of which have closed ends.

1/7

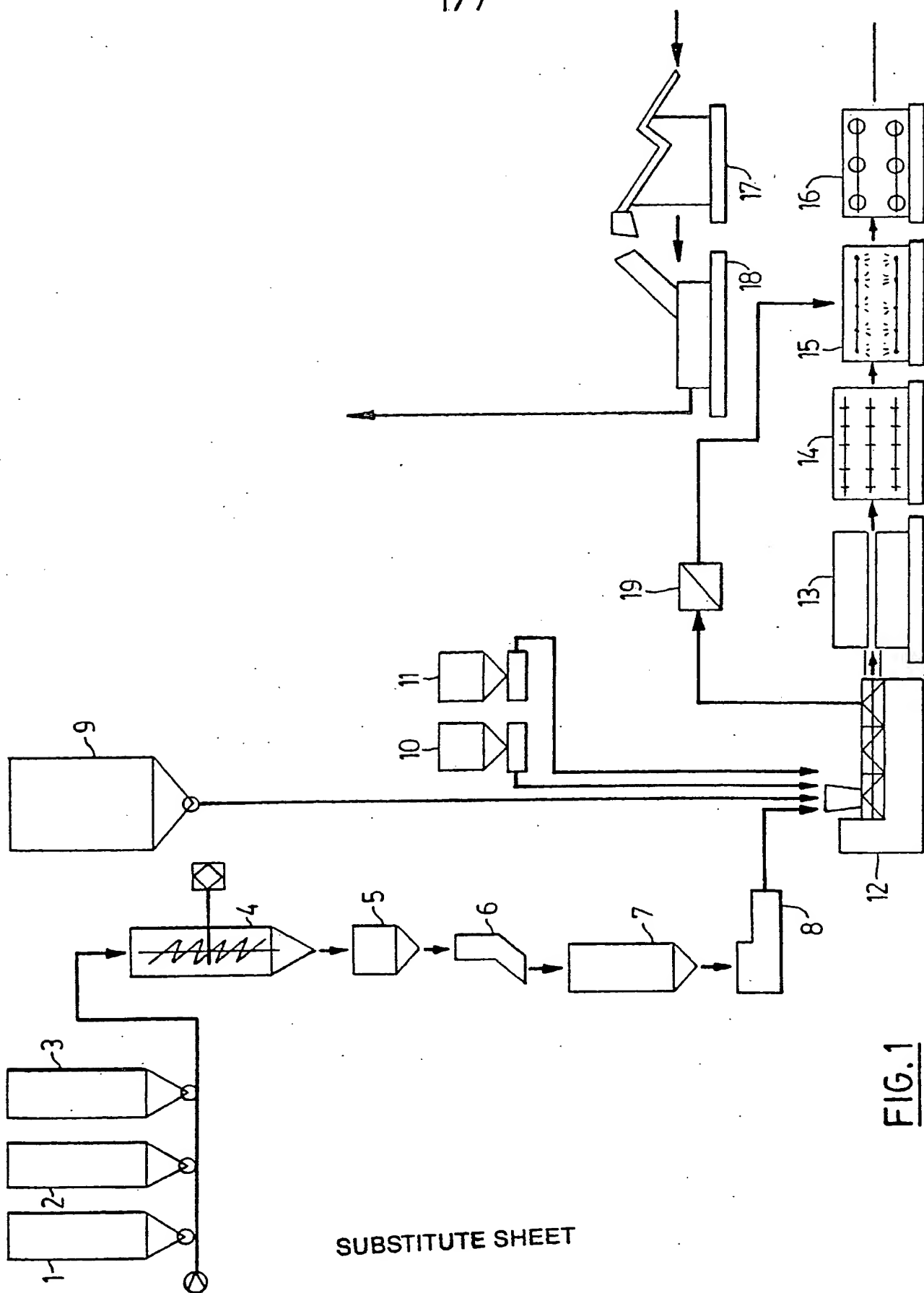
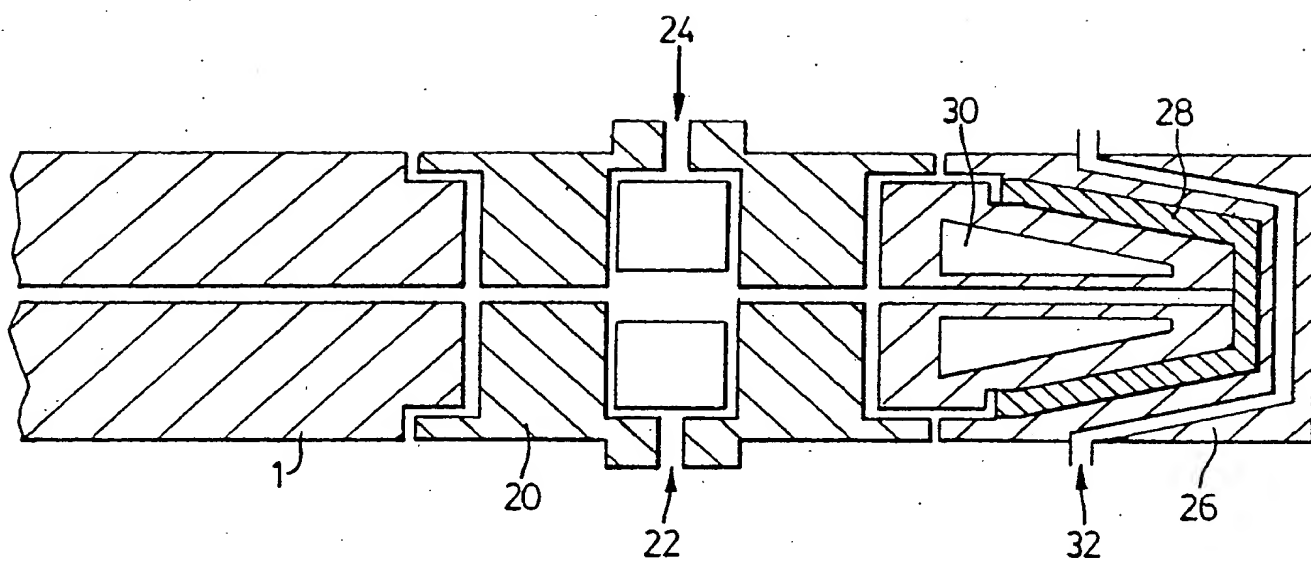


FIG. 1

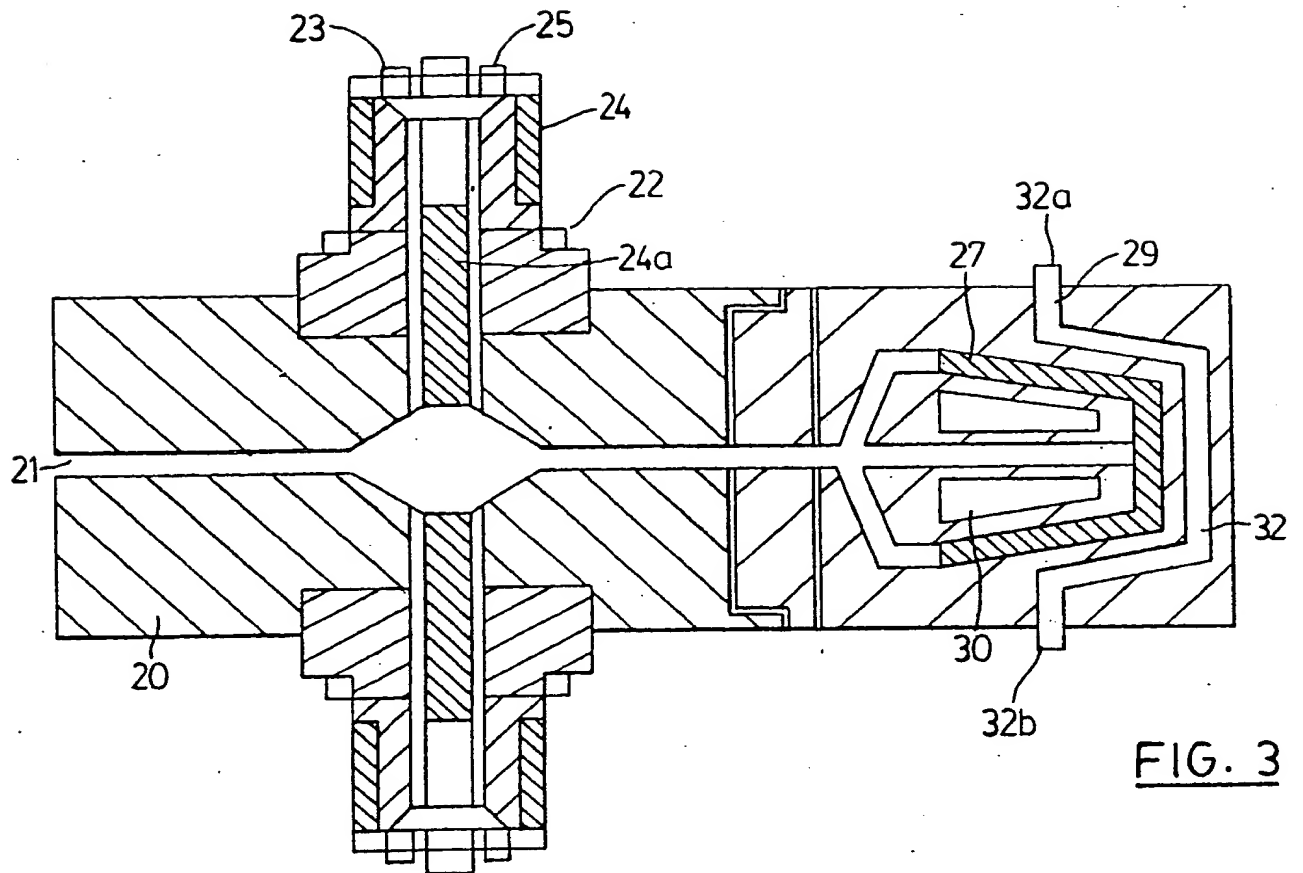
SUBSTITUTE SHEET

2/7

FIG. 2

SUBSTITUTE SHEET

3/7

FIG. 3

SUBSTITUTE SHEET

4/7

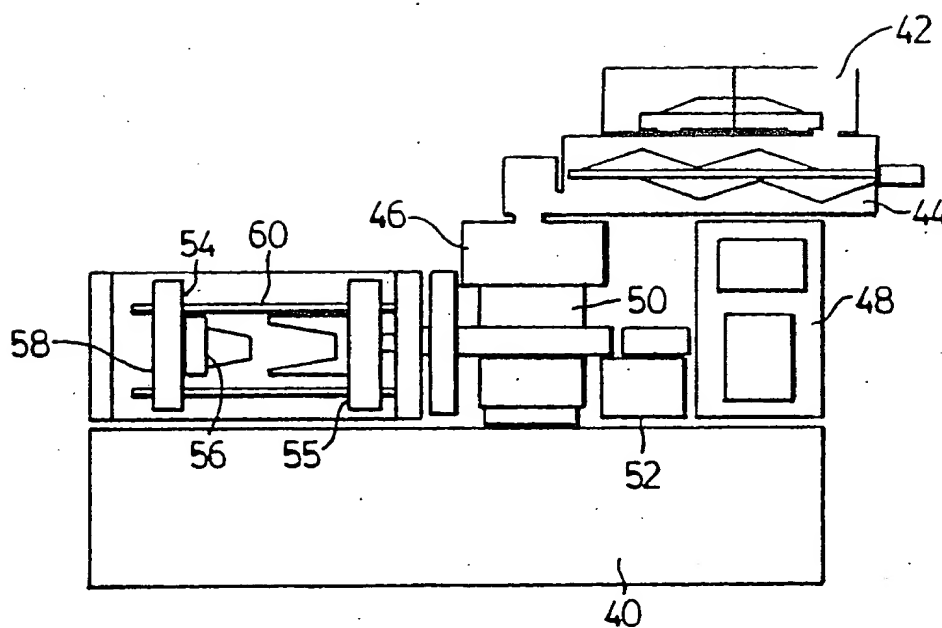


FIG. 4

SUBSTITUTE SHEET

5/7

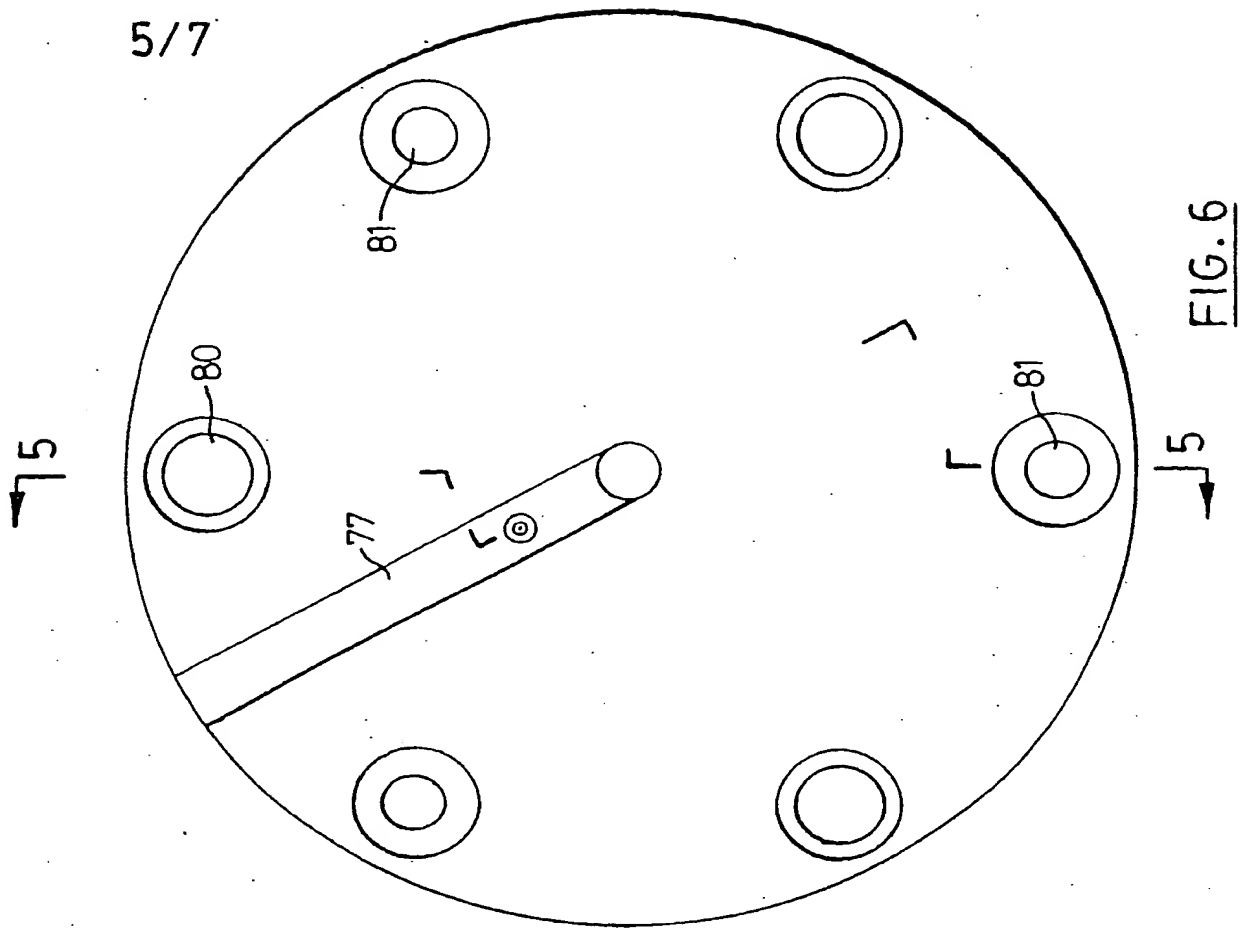


FIG. 6

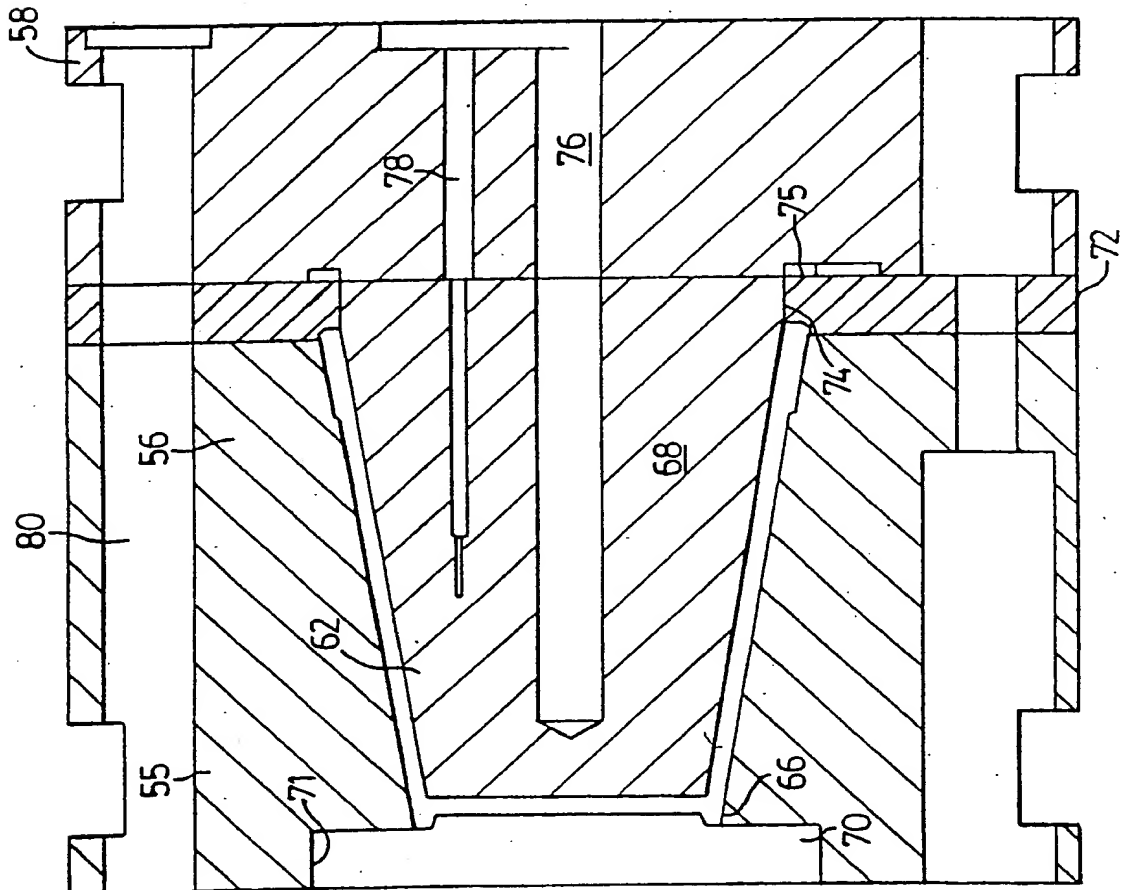


FIG. 5

SUBSTITUTE SHEET

6/7

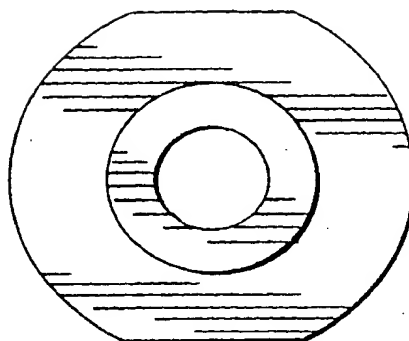


FIG. 7a

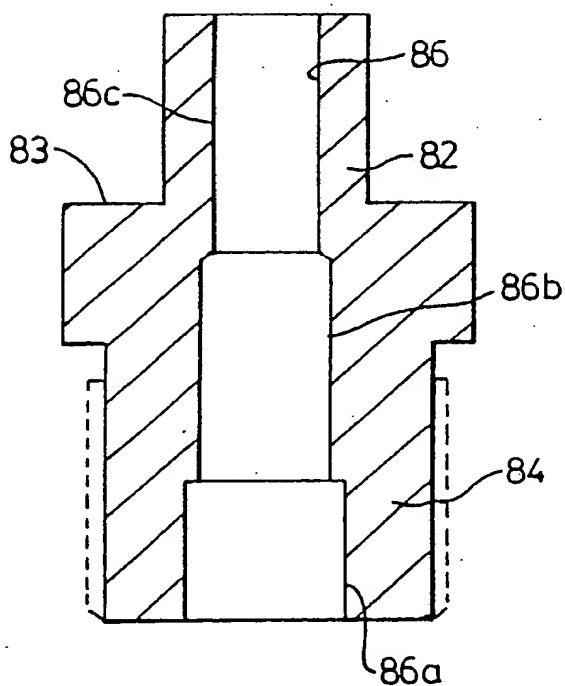


FIG. 7b

SUBSTITUTE SHEET

7/7

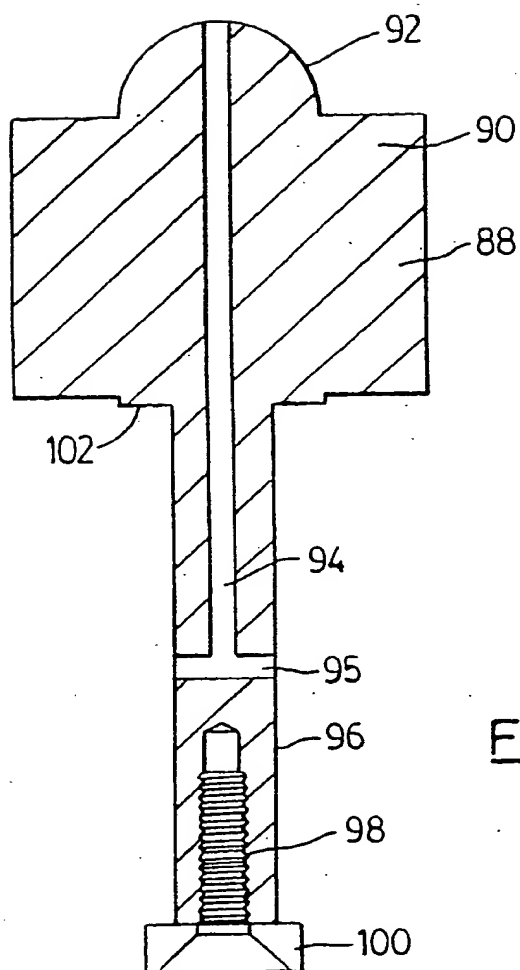


FIG. 8a

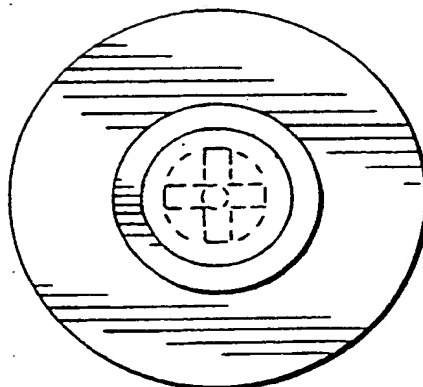


FIG. 8b

SUBSTITUTE SHEET

CORRECTED
VERSION*

PCT

WORLD INTELLECTUAL PROPERTY ORGANIZATION
International Bureau



INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification ⁵ : C08L 3/02, A23L 1/18 B65D 65/46, B29C 33/00	A3	(11) International Publication Number: WO 93/05668 (43) International Publication Date: 1 April 1993 (01.04.93)
(21) International Application Number: PCT/CA92/00406 (22) International Filing Date: 18 September 1992 (18.09.92) (30) Priority data: 761,221 19 September 1991 (19.09.91) US (60) Parent Application or Grant (63) Related by Continuation US 761,221 (CIP) Filed on 19 September 1991 (19.09.91) (71)(72) Applicant and Inventor: AUNG, David [CA/CA]; 7 Kimbercroft Court, Scarborough, Ontario M1S 4K7 (CA).		(74) Agent: BERESKIN & PARR; 40 King Street West, 40th Floor, Toronto, Ontario M5Y 3Y2 (CA). (81) Designated States: AT, AU, BB, BG, BR, CA, CH, CS, DE, DK, ES, FI, GB, HU, JP, KP, KR, LK, LU, MG, MN, MW, NL, NO, PL, RO, RU, SD, SE, US, Euro- pean patent (AT, BE, CH, DE, DK, ES, FR, GB, GR, IE, IT, LU, MC, NL, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, ML, MR, SN, TD, TG). Published <i>With international search report.</i> <i>Before the expiration of the time limit for amending the</i> <i>claims and to be republished in the event of the receipt of</i> <i>amendments.</i> (88) Date of publication of the international search report: 5 August 1993 (05.08.93)
(54) Title: MOULDING COMPOSITION, COMPRISING FLOUR, STARCH AND WATER, PROCESS FOR PREPA- RING THE SAME AND SHAPED ARTICLES THEREFROM (57) Abstract A composition suitable for forming into shaped articles comprising flour, starch, and water. The flour, starch and water are each present in an amount such that the composition is rigid and stable over a predetermined temperature range. The composition preferably contains 40 to 80 % by weight flour, 20-60 % starch and 15-25 % water and the flour and starch are obtained from natural cereal sources, such as corn, rice, potato, tapioca and wheat. A process of preparing the composition and a shaped article, and a shaped article produced in accordance with the process are also described. The shaped article may be used for foodstuffs.		

* (Referred to in PCT Gazette No. 19/1993, Section II)

FOR THE PURPOSES OF INFORMATION ONLY

Codes used to identify States party to the PCT on the front pages of pamphlets publishing international applications under the PCT.

AT	Austria	FR	France	MR	Mauritania
AU	Australia	GA	Gabon	MW	Malawi
BB	Barbados	GB	United Kingdom	NL	Netherlands
BE	Belgium	GN	Guinea	NO	Norway
BF	Burkina Faso	GR	Greece	NZ	New Zealand
BG	Bulgaria	HU	Hungary	PL	Poland
BJ	Benin	IE	Ireland	PT	Portugal
BR	Brazil	IT	Italy	RO	Romania
CA	Canada	JP	Japan	RU	Russian Federation
CF	Central African Republic	KP	Democratic People's Republic of Korea	SD	Sudan
CG	Congo	KR	Republic of Korea	SE	Sweden
CH	Switzerland	KZ	Kazakhstan	SK	Slovak Republic
CI	Côte d'Ivoire	LJ	Liechtenstein	SN	Senegal
CM	Cameroon	LK	Sri Lanka	SU	Soviet Union
CS	Czechoslovakia	LU	Luxembourg	TD	Chad
CZ	Czech Republic	MC	Monaco	TG	Togo
DE	Germany	MG	Madagascar	UA	Ukraine
DK	Denmark	MI	Mali	US	United States of America
ES	Spain	MN	Mongolia	VN	Viet Nam
FI	Finland				